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PLUTONIUM IN ATMOSPHERIC ENVIRONMENT

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Plutonium and its isotopes in the environment are concerned by public because of its chemical and radiological toxicity and fissile material. Researchers in the fields of atmospheric chemistry, chemical oceanography and others have been interesting in plutonium isotopes (^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu) in the environment as a unique transient tracer of atmospheric, oceanic, terrestrial and biogeochemical processes. Since explosions of the New Mexico and Nagasaki atomic bombs in 1945, global environment has contaminated with plutonium as a result of atmospheric nuclear weapons tests, satellite accidents and nuclear reactor accidents. Especially, large quantities of plutonium were released during atmospheric tests of nuclear weapons conducted by USA and former Soviet Union mainly during the 1950s and early 1960s. Atmospheric behaviour of plutonium has been frequently studied during the past 50 years. As a result, during the large-scale nuclear weapons tests of hydrogen bombs, radioactive debris including plutonium reached the stratosphere, which became then the main reservoir of plutonium. The stratospheric plutonium was transported into troposphere as an apparent stratospheric residence time of 1–2 years due to exchange processes between the stratosphere and the troposphere [1]. Although the stratospheric plutonium decreased to negligible level after 1990, plutonium has been detected in dust and deposition samples collected in Japan, Europe and US, in which current levels of $^{239, 240}$Pu are 0.1–10 nBq m^{-3}, 0.05–10 mBq m^{-2}Mon^{-1} for surface airborne dust and deposition, respectively. Sources of the atmospheric plutonium since 1990 are considered to be resuspension of deposited plutonium, including plutonium-bearing soil particles blew up by storms [2] and large-scale biomass burning.

Plutonium in environmental samples has been measured by alpha spectrometry. Recent development of mass spectrometric measurements such as ICP-MS, AMS and others allows us to determine ^{240}Pu/^{239}Pu and ^{241}Pu/^{239}Pu atom ratios in the environmental samples [3], which depend on scale of nuclear explosion and sources such as nuclear reactor accident. As results of measurement of archived samples, we have new knowledge about long-term variation of ^{240}Pu/^{239}Pu atom ratios in the atmospheric samples (deposition and dust). This knowledge is important to have better understanding of terrestrial and oceanic processes of plutonium.

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AMS AND ICP-MS FOR MEASUREMENT OF LOW LEVEL RADIONUCLIDES

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Keywords: AMS, ICP-MS, mass spectrometry, long-lived radionuclides, environmental radioactivity

Radionuclides are conventionally measured by detecting their characteristic radiation using alpha spectrometry, beta counting including liquid scintillation counting and gamma spectrometry depending on their decay modes, these methods are high sensitive for short half-lived radionuclides. Mass spectrometry, typically used to measure isotopes of elements, can be also used for measurement of radionuclides. In these methods, the atoms of the radionuclide of interest are directly measured. Therefore, mass spectrometry methods are normally sensitive for the measurement of

long-lived radionuclides. Among various inorganic mass spectrometric methods, inductively coupled plasma mass spectrometry (ICP-MS) and accelerator mass spectrometry (AMS) are two most popular used mass spectrometry techniques for the measurement of radionuclides, especially long-lived radionuclides. With the improvement of ICP-MS technique and more instruments to be installed, the application of this technique is becoming more popular tool for measurement of radionuclides. By hyphenation with automated separation system, ICP-MS will play a critical role in rapid determination of radionuclide for emergency analysis. AMS is the most sensitive analytical technique for many long-lived radionuclides, the new development of this techniques, especially the miniaturization of AMS system significantly reduce the cost of instrument as well as maintenance and operation, this stimulated and enhanced the application of this technique in the environmental researches. This work present the application of ICP-MS and AMS in the measurement of most important radionuclides, such as ^{99}Tc, ^{129}I, ^{135}Cs, ^{236}U, ^{237}Np, ^{239}Pu, ^{240}Pu, especially the new progress in the analytical methods of these radionuclides for environmental researches.

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FUKUSHIMA-DERIVED RADIOCESIUM IN THE WESTERN NORTH PACIFIC IN 2014

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Keywords: radiocesium, Fukushima Dai-ichi Nuclear Power Plant accident, North Pacific

Accident of Fukushima-Dai-ichi Nuclear Power Plant on 11 March 2011 resulted in a large amount release of radiocesium into the North Pacific Ocean. Since just after the accident, the Government of Japan, Tokyo Electric Power Co., and lots of oceanographers from countries of the North Pacific Rim have been measured concentration of dissolved radiocesium in seawater. They found that along surface currents between 40°N and 50°N approximately the released radiocesium had been transported eastward and reached the west coast of the North American Continent by April 2015. On the other hand, some of it were conveyed southward due to subduction of the subtropical mode water (STMW). Maximum of radiocesium concentration in subsurface layer (200–400 m depth), which is derived from the subduction of STMW, should be spreading in the subtropical area of the western North Pacific during the past four years. However, behaviour of the subsurface maximum is still unknown. We collected seawater samples from surface to about 800 m depth at 14 stations between 15°N and 41°N in the western North Pacific in 2014 and revealed vertical profiles of Fukushima-derived radiocesium at each station. At a station (34°N/148°E) in the just south of the Kuroshio Extension, where STMW was formed just after the accident, activity concentration of ^{137}Cs in the subsurface layer has been decreasing; about 50 Bq/m^3 in October 2011, 20 Bq/m^3 in January 2012, 10 Bq/m^3 in November 2012, and 5 Bq/m^3 in July 2014. At a station (30°N/148°E) located about 400 km directly south of the above station, the subsurface activity concentration increased from about 8 Bq/m^3 in January 2012 to 15 Bq/m^3 in November 2012. Then it has fallen to about 5 Bq/m^3 by July 2014. At a station (22°N/165°E) further south, the activity concentration in the subsurface layer rose from about 2 to 5 Bq/m^3 between June 2012 and December 2014. These temporal changes in the activity concentration of ^{137}Cs suggest that Fukushima-derived radiocesium has been spreading southward through the subsurface layer along circulation of STMW in the subtropical area.